

Application No.:09/822,531
Amendment dated: September 2, 2003
Reply to Office Action of May, 30, 2003

Amendments to the Claims:

This listing of claims will replace all prior versions, and listings of claims in the application.

Listing of Claims:

Claim 1 (currently amended): A method for producing aromatic carbonates which comprises the steps of :

(i) contacting at a temperature sufficient to keep the mixture molten at least one aromatic hydroxy compound with a catalyst composition ~~comprising~~ consisting essentially of the following and any reaction products thereof:

(A) at least one Group 8, 9, or 10 metal or a compound thereof;

(B) at least one tetrafluoroborate, hexafluorophosphate, tetraarylborate, arylsulfonate, sulfate, nitrate, carboxylate, acetate, benzoate, halide, chloride, or bromide salt with cation selected from the group consisting of guanidinium, ammonium, phosphonium, sulfonium, and alkali metal;

(C) at least one metal co-catalyst; and

(D) optionally, at least one activating solvent;

(ii) optionally heating the mixture at atmospheric pressure to a temperature above that sufficient to keep the mixture molten;

(iii) pressurizing the mixture with carbon monoxide;

(iv) optionally heating the mixture under pressure of carbon monoxide to a temperature above that sufficient to keep the mixture molten;

(v) optionally maintaining the mixture under pressure of carbon monoxide for a time period;

(vi) introducing oxygen to the mixture to a desired concentration of oxygen in carbon monoxide with the proviso that the solvent for the reaction mixture before introduction of oxygen consists of the aromatic hydroxy compound;

(vii) starting gas flow to the mixture at a desired concentration of oxygen and carbon monoxide;

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(viii) optionally maintaining gas flow for a time period at less than a desired ultimate temperature for the mixture; and

(ix) optionally heating the mixture to a desired ultimate temperature under flow of gases.

Claim 2 (original): The method of claim 1 wherein the aromatic hydroxy compound is phenol.

Claim 3 (original): The method of claim 1 wherein the at least one Group 8, 9, or 10 metal is a palladium source.

Claim 4 (cancelled)

Claim 5 (cancelled)

Claim 6 (previously presented): The method of claim 1 wherein the salt is at least one quaternary ammonium salt.

Claim 7 (original): The method of claim 6 wherein the salt is at least one bromide or chloride salt with cation selected from the group consisting of tetraethylammonium, tetramethylammonium, tetrabutylammonium, and methyltributylammonium.

Claim 8 (previously presented): The method of claim 1 wherein the salt is at least one alkali metal salt and the mixture contains at least one activating solvent selected from the group consisting of polyethers, nitriles, carboxylic acid amides, and sulfones.

Claim 9 (original): The method of claim 8 wherein the alkali metal salt is sodium bromide.

Claim 10 (original): The method of claim 8 wherein the activating solvent is at least one polyether.

Claim 11 (original): The method of claim 1 wherein the metal co-catalyst is selected from the group consisting of lead, copper, titanium, cobalt, manganese, zinc, bismuth, zirconium, tungsten, chromium, nickel, iron, lanthanide metals, cerium, and ytterbium.

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and mixtures thereof.

Claim 12 (original): The method of claim 11 wherein the metal co-catalyst is selected from the group consisting of lead, cobalt, copper, titanium, manganese, cerium, and mixtures thereof.

Claim 13 (original): The method of claim 1 wherein the composition further comprises a base.

Claim 14 (original): The method of claim 13 wherein the base is at least one of sodium hydroxide or sodium phenoxide.

Claim 15 (original): The method of claim 1 wherein the mixture is heated in step (ii) at atmospheric pressure to a temperature no higher than about 90°C.

Claim 16 (original): The method of claim 15 wherein the mixture is heated in step (ii) at atmospheric pressure to a temperature in a range of between about 72°C and about 90°C.

Claim 17 (original): The method of claim 16 wherein the mixture is held under pressure of carbon monoxide in step (v) for a time period in a range of between about 5 minutes and about 40 minutes.

Claim 18 (original): The method of claim 17 wherein oxygen is introduced to the mixture in step (vi) at a concentration greater than about 8 molar % based on carbon monoxide.

Claim 19 (original): The method of claim 18 wherein gas flow is maintained for a time period at less than a desired ultimate temperature for the mixture in step (viii) and the mixture is heated to a desired ultimate temperature under flow of gases in step (ix).

Claim 20 (original): The method of claim 1 wherein the mixture is heated in step (ii) at atmospheric pressure to a temperature no higher than about 72°C.

Claim 21 (original): The method of claim 20 wherein oxygen is introduced to the mixture in step (vi) at a concentration in a range of between about 5.5 molar % and about 9.5 molar % based on carbon monoxide.

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Claim 22 (original): The method of claim 21 wherein the mixture is heated to a desired ultimate temperature under flow of gases in step (ix).

Claim 23 (original): The method of claim 20 wherein the mixture is held under pressure of carbon monoxide in step (v) for a time before introducing oxygen to the mixture.

Claim 24 (original): The method of claim 23 wherein oxygen is introduced to the mixture in step (vi) at a concentration greater than about 8 molar % based on carbon monoxide.

Claim 25 (original): The method of claim 24 wherein the mixture is heated to a desired ultimate temperature under flow of gases in step (ix).

Claim 26 (original): The method of claim 1 wherein the mixture is heated in step (iv) under pressure of carbon monoxide to a desired ultimate temperature.

Claim 27 (original): The method of claim 26 wherein the mixture is held under pressure of carbon monoxide in step (v) for a time before introducing oxygen to the mixture.

Claim 28 (original): The method of claim 27 wherein oxygen is introduced to the mixture in step (vi) at a concentration greater than about 8 molar % based on carbon monoxide.

Claim 29 (original): The method of claim 1 wherein the mixture is heated in step (iv) under pressure of carbon monoxide to a temperature above that sufficient to keep the mixture molten and below a desired ultimate reaction temperature.

Claim 30 (original): The method of claim 29 wherein the mixture is held under pressure of carbon monoxide in step (v) for a time before introducing oxygen to the mixture.

Claim 31 (original): The method of claim 30 wherein oxygen is introduced to the mixture in step (vi) at a concentration greater than about 8 molar % based on carbon monoxide.

Claim 32 (original): The method of claim 31 wherein the mixture is heated to a desired ultimate temperature under flow of gases in step (ix).

Claim 33 (currently amended) A method for producing diphenyl carbonate which

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comprises the steps of :

(i) contacting phenol at a temperature sufficient to keep the mixture molten with a catalyst composition ~~comprising~~ consisting essentially of the following and any reaction products thereof:

(A) at least one palladium source;

(B) at least one tetrafluoroborate, hexafluorophosphate, tetraarylborate, arylsulfonate, sulfate, nitrate, carboxylate, acetate, benzoate, halide, chloride, or bromide salt with cation selected from the group consisting of guanidinium, ammonium, phosphonium, sulfonium, and alkali metal;

(C) at least one metal co-catalyst with metal selected from the group consisting of lead, cobalt, copper, titanium, manganese, cerium, and mixtures thereof; and

(D) optionally, at least one activating solvent;

(ii) heating the mixture at atmospheric pressure to a temperature in a range of between about 72°C and about 90°C;

(iii) pressurizing the mixture with carbon monoxide;

(v) maintaining the mixture under pressure of carbon monoxide for a time period;

(vi) introducing oxygen to the mixture at a concentration of greater than about 8 molar % based on carbon monoxide;

(vii) starting gas flow to the mixture at a desired concentration of oxygen and carbon monoxide;

(viii) maintaining gas flow for a time period at less than a desired ultimate temperature for the mixture; and

(ix) heating the mixture to a desired ultimate temperature under flow of gases.

Claim 34 (currently amended) A method for producing diphenyl carbonate which comprises the steps of :

(i) contacting phenol at a temperature sufficient to keep the mixture molten with a catalyst composition ~~comprising~~ consisting essentially of the following and any reaction products thereof:

(A) at least one palladium source;

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(B) at least one tetrafluoroborate, hexafluorophosphate, tetraarylborate, arylsulfonate, sulfate, nitrate, carboxylate, acetate, benzoate, halide, chloride, or bromide salt with cation selected from the group consisting of guanidinium, ammonium, phosphonium, sulfonium, and alkali metal;

(C) at least one metal co-catalyst with metal selected from the group consisting of lead, cobalt, copper, titanium, manganese, cerium, and mixtures thereof; and

(D) optionally, at least one activating solvent;

(iii) pressurizing the mixture with carbon monoxide;

(iv) heating the mixture under pressure of carbon monoxide to a desired ultimate temperature;

(v) maintaining the mixture under pressure of carbon monoxide for a time period;

(vi) introducing oxygen to the mixture at a concentration of greater than about 8 molar % based on carbon monoxide; and

(vii) starting gas flow to the mixture at a desired concentration of oxygen and carbon monoxide.

Claim 35 (currently amended): A method for producing diphenyl carbonate which comprises the steps of :

(i) contacting phenol at a temperature sufficient to keep the mixture molten with a catalyst composition ~~comprising~~ consisting essentially of the following and any reaction products thereof:

(A) at least one palladium source;

(B) at least one tetrafluoroborate, hexafluorophosphate, tetraarylborate, arylsulfonate, sulfate, nitrate, carboxylate, acetate, benzoate, halide, chloride, or bromide salt with cation selected from the group consisting of guanidinium, ammonium, phosphonium, sulfonium, and alkali metal;

(C) at least one metal co-catalyst with metal selected from the group consisting of lead, cobalt, copper, titanium, manganese, cerium, and mixtures thereof; and

(D) optionally, at least one activating solvent;

(ii) heating the mixture at atmospheric pressure to a temperature no higher than about 72°C;

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- (iii) pressurizing the mixture with carbon monoxide;
- (vi) introducing oxygen to the mixture to a desired concentration of oxygen in carbon monoxide;
- (vii) starting gas flow to the mixture at a desired concentration of oxygen and carbon monoxide; and
- (ix) heating the mixture to a desired ultimate temperature under flow of gases.

Claim 36 (currently amended): A method for producing diphenyl carbonate which comprises the steps of :

(i) contacting phenol at a temperature sufficient to keep the mixture molten with a catalyst composition ~~comprising~~ consisting essentially of the following and any reaction products thereof:

- (A) at least one palladium source;
- (B) at least one tetrafluoroborate, hexafluorophosphate, tetraarylborate, arylsulfonate, sulfate, nitrate, carboxylate, acetate, benzoate, halide, chloride, or bromide salt with cation selected from the group consisting of guanidinium, ammonium, phosphonium, sulfonium, and alkali metal;
- (C) at least one metal co-catalyst with metal selected from the group consisting of lead, cobalt, copper, titanium, manganese, cerium, and mixtures thereof; and
- (D) optionally, at least one activating solvent;

- (ii) heating the mixture at atmospheric pressure to a temperature no higher than about 72°C;
- (iii) pressurizing the mixture with carbon monoxide;
- (v) maintaining the mixture under pressure of carbon monoxide for a time period;
- (vi) introducing oxygen to the mixture at a concentration of greater than about 8 molar % based on carbon monoxide;
- (vii) starting gas flow to the mixture at a desired concentration of oxygen and carbon monoxide; and
- (ix) heating the mixture to a desired ultimate temperature under flow of gases.

Claim 37 (currently amended): A method for producing diphenyl carbonate which

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comprises the steps of :

(i) contacting phenol at a temperature sufficient to keep the mixture molten with a catalyst composition ~~comprising~~ consisting essentially of the following and any reaction products thereof:

(A) at least one palladium source;

(B) at least one tetrafluoroborate, hexafluorophosphate, tetraarylborate, arylsulfonate, sulfate, nitrate, carboxylate, acetate, benzoate, halide, chloride, or bromide salt with cation selected from the group consisting of guanidinium, ammonium, phosphonium, sulfonium, and alkali metal;

(C) at least one metal co-catalyst with metal selected from the group consisting of lead, cobalt, copper, titanium, manganese, cerium, and mixtures thereof; and

(D) optionally, at least one activating solvent;

(iii) pressurizing the mixture with carbon monoxide;

(iv) heating the mixture under pressure of carbon monoxide to temperature above that sufficient to keep the mixture molten and below a desired ultimate reaction temperature;

(v) maintaining the mixture under pressure of carbon monoxide for a time period;

(vi) introducing oxygen to the mixture at a concentration of greater than about 8 molar % based on carbon monoxide;

(vii) starting gas flow to the mixture at a desired concentration of oxygen and carbon monoxide; and

(ix) heating the mixture to a desired ultimate temperature under flow of gases.

Claim 38 (currently amended): A method for producing diphenyl carbonate which comprises the steps of :

(i) contacting phenol at a temperature sufficient to keep the mixture molten with a catalyst composition ~~comprising~~ consisting essentially of the following and any reaction products thereof:

(A) at least one palladium source;

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(B) at least one tetrafluoroborate, hexafluorophosphate, tetraarylborate, arylsulfonate, sulfate, nitrate, carboxylate, acetate, benzoate, halide, chloride, or bromide salt with cation selected from the group consisting of guanidinium, ammonium, phosphonium, sulfonium, and alkali metal;

(C) at least one metal co-catalyst with metal selected from the group consisting of lead, cobalt, copper, titanium, manganese, cerium, and mixtures thereof; and

(D) optionally, at least one activating solvent;

(iii) pressurizing the mixture with carbon monoxide;

(vi) introducing oxygen to the mixture at a concentration of greater than about 8 molar % based on carbon monoxide with the proviso that the solvent for the reaction mixture before the introduction of oxygen consists of the aromatic hydroxy compound;

(vii) starting gas flow to the mixture at a desired concentration of oxygen and carbon monoxide; and

(ix) heating the mixture to a desired ultimate temperature under flow of gases.

Claim 39 (currently amended): A method for producing aromatic carbonates which comprises the steps of :

(i) contacting at a temperature sufficient to keep the mixture molten at least one aromatic hydroxy compound with a catalyst composition ~~comprising~~ consisting essentially of the following and any reaction products thereof:

(A) at least one Group 8, 9, or 10 metal or a compound thereof;

(B) at least one tetrafluoroborate, hexafluorophosphate, tetraarylborate, arylsulfonate, sulfate, nitrate, carboxylate, acetate, benzoate, halide, chloride, or bromide salt with cation selected from the group consisting of guanidinium, ammonium, phosphonium, sulfonium, and alkali metal;

(C) at least one metal co-catalyst; and

(D) optionally, at least one activating solvent;

wherein the mixture is exposed to an atmosphere comprising at least about 8 molar % oxygen before the mixture is heated to a temperature above about 90°C, with the proviso that the solvent for the reaction mixture before the introduction of oxygen consists of the aromatic hydroxy compound.

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Claim 40 (original): The method of claim 39 wherein the aromatic hydroxy compound is phenol.

Claim 41 (original): The method of claim 39 wherein the at least one Group 8, 9, or 10 metal is a palladium source.

Claim 42 (currently amended): A method for producing aromatic carbonate from a mixture ~~comprising~~ consisting essentially of an aromatic hydroxy compound, which comprises the steps of :

(x) maintaining the mixture at a temperature at least sufficient to keep the mixture molten;

(xi) introducing oxygen and carbon monoxide to the mixture to a desired pressure with the proviso that the solvent for the reaction mixture before the introduction of oxygen consists of the aromatic hydroxy compound;

(xii) starting gas flow to the mixture at a desired concentration of oxygen and carbon monoxide;

(xiii) heating the mixture to a temperature in a range between that sufficient to keep the mixture molten and a desired ultimate temperature; and

(xiv) contacting the mixture with one or more mixtures ~~comprising~~ consisting essentially of an aromatic hydroxy compound and one or more catalyst components comprising the following and any reaction products thereof:

(A) at least one Group 8, 9, or 10 metal or a compound thereof;

(B) at least one tetrafluoroborate, hexafluorophosphate, tetraarylborate, arylsulfonate, sulfate, nitrate, carboxylate, acetate, benzoate, halide, chloride, or bromide salt with cation selected from the group consisting of guanidinium, ammonium, phosphonium, sulfonium, and alkali metal;

(C) at least one metal co-catalyst; and

(D) optionally, at least one activating solvent.

Claim 43 (original): The method of claim 42 wherein the aromatic hydroxy compound is phenol.

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Claim 44 (original): The method of claim 42 wherein the at least one Group 8, 9, or 10 metal is a palladium source.

Claim 45-46 (canceled)

Claim 47 (previously presented): The method of claim 42 wherein the salt is at least one quaternary ammonium salt.

Claim 48 (original): The method of claim 47 wherein the salt is at least one bromide or chloride salt with cation selected from the group consisting of tetraethylammonium, tetramethylammonium, tetrabutylammonium, and methyltributylammonium.

Claim 49 (previously presented): The method of claim 42 wherein the salt is at least one alkali metal salt and the mixture contains at least one activating solvent selected from the group consisting of polyethers, nitriles, carboxylic acid amides, and sulfones.

Claim 50 (original): The method of claim 49 wherein the alkali metal salt is sodium bromide.

Claim 51 (original): The method of claim 49 wherein the activating solvent is at least one polyether.

Claim 52 (original): The method of claim 42 wherein the metal co-catalyst is selected from the group consisting of lead, copper, titanium, cobalt, manganese, zinc, bismuth, zirconium, tungsten, chromium, nickel, iron, lanthanide metals, cerium, and ytterbium, and mixtures thereof.

Claim 53 (original): The method of claim 52 wherein the metal co-catalyst is selected from the group consisting of lead, cobalt, copper, titanium, manganese, cerium, and mixtures thereof.

Claim 54 (original): The method of claim 42 wherein the composition further comprises a base.

Claim 55 (original): The method of claim 54 wherein the base is at least one of sodium

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hydroxide or sodium phenoxide.

Claim 56 (currently amended): A method for producing diphenyl carbonate from a mixture comprising consisting essentially of phenol, which comprises the steps of :

(x) maintaining the mixture at a temperature at least sufficient to keep the mixture molten;

(xi) introducing oxygen and carbon monoxide to the mixture to a desired pressure with the proviso that the solvent for the reaction mixture before introduction of oxygen consists of the aromatic hydroxy compound;

(xii) starting gas flow to the mixture at a desired concentration of oxygen and carbon monoxide;

(xiii) heating the mixture to a temperature in a range between that sufficient to keep the mixture molten and a desired ultimate temperature; and

(xiv) contacting the mixture with one or more mixtures comprising phenol and one or more catalyst components comprising the following and any reaction products thereof:

(A) at least one palladium source;

(B) at least one tetrafluoroborate, hexafluorophosphate, tetraarylborate, arylsulfonate, sulfate, nitrate, carboxylate, acetate, benzoate, halide, chloride, or bromide salt with cation selected from the group consisting of guanidinium, ammonium, phosphonium, sulfonium, and alkali metal;

(C) at least one metal co-catalyst with metal selected from the group consisting of lead, cobalt, copper, titanium, manganese, cerium, and mixtures thereof; and

(D) optionally, at least one activating solvent.